

July 19, 2000

Mr. Ron Pauer
Lawrence Berkeley National Laboratory
1 Cyclotron Rd. – 75B
Berkeley, CA 94720

Dear Ron:

Here are my initial comments on the Preliminary Technical Report by Bernd Franke and Anthony Greenhouse who prepared an independent review of “Radiological Monitoring at LBNL” through a contract with the City of Berkeley.

As I have only recently received a copy of their preliminary report, I have not had the opportunity to perform an in-depth evaluation of each comment given by Franke and Greenhouse. Nevertheless, upon initial reading, I find the review to be thorough and objective. Some concerns are raised in this report that should lead to further enhancement of the laboratory's environmental monitoring program.

The review by Franke and Greenhouse is a preliminary analysis. Some of their statements may be revised and/or retracted pending further evaluation of information. I therefore recommend that each issue identified in their preliminary report be systematically reviewed in detail and that a response by LBNL be forwarded to the City of Berkeley.

In this correspondence, I shall offer my own comments to each of the issues raised in the Executive Summary of the June 30, 2000 Preliminary Technical Report by Franke and Greenhouse. Please note that my present remarks may also be modified in the future reflecting additional time to consider the Franke and Greenhouse preliminary findings and recommendations in greater depth.

Franke and Greenhouse Comment No. 1.

Ambient air monitoring for tritium should be expanded. The number of monitoring sites at LBNL is well below the DOE average. At other DOE facilities with similar amounts of tritium emissions at least 16 wind directions (of 22.5 degrees) are monitored. A comparable network would be advisable for LBNL as well.

The recommendation for expanding the ambient air monitoring network should be given serious consideration. However, I recommend that the technical basis for each additional air monitoring station be clearly established before siting any new stations. The operations at the NTLF cannot be compared with those at other DOE sites. The scale of operations potentially generating releases of radioactivity at most other DOE sites is substantially larger than those of the NTLF at LBNL. Other DOE sites, like the Savannah River Plant, have multiple facilities and sources of operation, each of which may release radioactivity to the atmosphere. These individual facilities are usually spread over a very large area; thus a larger network of ambient air monitors is indeed required to characterize environmental concentrations resulting from multiple onsite operations that change as the result of changes in the DOE mission at these sites.

For example, many of the DOE sites indicated in Table 3 of the review by Franke and Greenhouse have substantial tritium inventories that exceed the amounts present at the NTLF by orders of magnitude. Other facilities such as LLNL, have reduced their tritium inventory recently, but future increases are anticipated. They therefore continue to use their historical monitoring network to avoid the expense associated with decommissioning and then re-commissioning the network to coincide with fluctuations in future tritium operations.

LLNL has two sites; Main and Site 300. In addition, they have several significant diffuse sources on their Main Site. If one looks at only perimeter and remote locations for the Main Site, the total was 13 for 1998 and not 20 (LLNL SER, 1999). The other DOE sites identified may also have similar situations or conditions.

The technical basis for siting new monitoring stations should be thoroughly developed to ensure there is sufficient value for the information obtained. The placement of monitoring locations is greatly influenced by the topography of LBNL. Much more channeling of wind occurs than for flat land terrain like Savannah River and Pantex. During the 1990's, Berkeley Lab operated several off-site ambient air sampling systems that monitored tritium. These systems were eventually discontinued because data from those locations primarily confirmed that tritium from NTLF were either below limits of detection or not distinguishable from background. The results of these monitoring stations have been published in LBNL annual Site Environmental Reports. These reports have been given to Franke and Greenhouse.

Present ambient air monitors as well as two different mathematical models of air dispersion show that exposures have been far below regulatory limits (as much as a factor of 100 below EPA's National Environmental Standards for Hazardous Air Pollutants, NESHAPS, which for radioactivity is set at 10 mrem/y). The most recent monitor added to LBNL's network of monitoring stations was placed onsite, in the primary wind direction 21 meters from the stack (in the eucalyptus grove with an intake at stack height). Readings from this monitor, to date, indicate

that, even under the hypothetical condition that someone resided full time at that location, exposures would be more than a factor of 10 below the NESHAP standard. Thus, off-site short-term exposures in wind directions other than those selected to define the maximally exposed individual, should be much lower than 1% of the NESHAP standard. Additional assessments of the downwind dispersion of short-term emissions of tritium from the NTLF should be undertaken to provide insights as to the location and extent of potential employee and public exposures to such episodes.

Franke and Greenhouse Comment No. 2.

Releases of tritium are often in short bursts (e.g. 0.2 Ci of tritium emitted over 17 minutes on March 28, 1998). This renders the results of the computer program used to determine compliance with the NESHAP standard (CAP88PC) to be inaccurate. The probability that a person near the fence could receive a radiation dose of greater than 10 mrem/yr should be determined using appropriate models, accounting for the complex terrain and the discontinuous nature of the releases. However, there is no evidence at this time to suggest that offsite exposures resulted in radiation doses exceeding the 10 mrem/yr limit for any individual.

Our most recent application of the CALPUFF model accounts for complex terrain and the discontinuous nature of releases. I concur with Franke and Greenhouse: there is no evidence at this time to suggest that the 10 mrem per year regulatory limit has been exceeded by any individual residing beyond the site boundary of LBNL. This conclusion is based on both modeling of stack releases and the use of ambient environmental measurements, including limited bioassay measurements of individuals on and offsite. Additional conservatism is obtained through LBNL's use of the conservative assumption of adding releases of tritiated hydrogen gas (HT) to releases of tritiated water vapor (HTO) and treating the sum as 100% HTO. In the environment, an exposure to HT produce doses that are 10,000 times lower than a similar exposure to HTO.

To date, the shortest time periods of tritium releases that have been evaluated are monthly averages. Comparison of CALPUFF results to those produced with the EPA mandated model, CAP 88 PC, indicates a general tendency for CAP 88 PC to overestimate measured concentrations. In general, CAP 88 PC produced higher concentrations, by a factor of 2 to 10, than those predicted with the more sophisticated CALPUFF modeling approach. Thus, changing from CAP 88 PC to CALPUFF is likely to produce even lower values of offsite exposure.

We are currently in the process of using the CALPUFF model to evaluate maximum and average exposures that could result from the release of tritium over time periods as short as 15 minutes to one hour or more. More detailed information on our results of this analysis should be available in time for the August 10th Public Sampling Task Force Meeting in Berkeley.

Franke and Greenhouse Comment No. 3.

There are minor uncertainties associated with the measurements of tritium in ambient air at a given location. Some corrections are necessary.

Minor uncertainties are to be expected when making measurements of natural systems. It is more important to determine if there is a systematic bias versus a random error. Random errors are less important when calculating average values. Franke and Greenhouse suggest that information regarding the uncertainty of analytical data be incorporated in the Site Environmental Reports. I agree with this recommendation. This should include general statements about the uncertainty in ambient air measurements and specific uncertainty values for each reported sampling result. In general, however, the uncertainty associated with the analytical data for HTO in air samples is reasonable.

In discussing the ratio $(\text{HTO}+\text{HT})/\text{HTO}$ of stack monitoring results, Franke seems to be referring to the inverse, i.e. $\text{HTO}/(\text{HTO}+\text{HT})$, which should be less than 1.2. Perhaps this is a typo in the IFEU report.

Franke and Greenhouse Comment No. 4.

Tritium inventory data is uncertain in the order of +/- 30% and not suitable to verify airborne releases. However, improved inventory data is useful to verify the type of operations at NTLF.

This conclusion by Franke and Greenhouse supports numerous statements made by LBNL staff in public meetings. The uncertainty in the present tritium inventory data is not suitable for performing a mass balance calculation in order to verify releases from the NTLF. Releases from the NTLF are most reliably indicated by real time sampling at the stack and environmental sampling for concentrations of HTO in ambient air, which are routinely performed by LBNL. The data collected from real time and composite stack sampling and ambient air monitoring data are integrated to obtain a thorough record of NTLF releases to the environment. However, to improve the tracking of tritium storage at the NTLF, LBNL has committed to purchase a calorimeter that is especially designed to account for the tritium inventory with much greater precision than was possible in the past.

On page 6 of the report by Franke and Greenhouse, I note some errors of fact. Their report says, "...all numbers are rounded to 0.1 g of tritium, or 96 Ci; the reporting threshold is 0.05 g of tritium of 48 Ci." One gram of pure tritium gas is 9600 Ci; therefore 0.1g is 960 Ci and 0.05 g is 480 Ci. On the same page, Franke and Greenhouse state that "...shipments up until 1991 were typically between 100 and 300 Ci per year". To my knowledge, NTLF product shipments rarely exceeded 1 Ci and usually were in the range of 0.1 to 100 millicuries. Even if we assumed an average shipment activity of 0.1 Ci and a shipment frequency of 100 per year, the product shipment of total tritium would only be 10 Ci. Franke and Greenhouse may be considering non-product shipments such as the return of tritium contaminated shipping containers to the supplier.

Franke and Greenhouse Comment. No. 5.

Historical data for tritium in ambient air is somewhat puzzling. Concentrations in ambient air do not correlate with reported releases, maximum levels were reported for Building 3 (Calvin) in

1977/1978. The integrated concentrations of tritium are similar to those at NTLF indicating the possibility that contamination of soil and groundwater may have occurred there as well. A preliminary sampling effort around the building is recommended.

I am in the process of reviewing the reasons for the discrepancies reported between 1977 and 1978 for Building 3 (Calvin) at the University of California. The quality of environmental monitoring at LBNL has been upgraded substantially from the techniques that were used prior to 1985. The improvements in the reliability of environmental monitoring both at the stack and in the environment explain most of the discrepancies indicated by Franke and Greenhouse. The discrepancies identified by Franke and Greenhouse appear to occur prior to 1987. They appear most pronounced prior to 1980.

Some of this discrepancy appears to be due to the fact that the air sampler was placed in close proximity to the roof vent of the building and is not representative of exposures to individuals on or off site. As such, the HTO levels reported for Building 3 are not directly comparable with those reported for offsite monitoring stations. The offsite monitoring stations are placed in those locations specifically for the purpose of detecting concentrations that could lead to maximum exposures to members of the public.

In addition, the historical data from the Calvin Laboratory location (and other locations during that time period) were generated with an old sampling system and analytical procedure. Much of the data (yearly averages) from this site may appear to be elevated because of the higher Minimum Detectable Activity (MDA) associated with the monitoring method (sampling and analysis). Preliminary examination of the data suggests that the nominal MDA at that time was on the order of 100's of pCi per cubic meter.

The historical MDA for those samples should be determined to evaluate the extent to which this issue is also partially responsible for the discrepancies reported by Franke and Greenhouse. These data are still in the process of being reviewed and additional comments will be forthcoming in the near future. For perspective, the current contract LBNL has with its commercial laboratory requires that all analyses meet a MDA of 10 pCi per cubic meter. On average, the reported MDA is between 3 and 4 pCi per cubic meter.

Franke and Greenhouse Comment No. 6.

With regard to historical radiation exposures, gamma and neutron doses are of the greatest concern. Levels at Olympus Gate may have exceeded then-prevailing dose limits in 1959 and 1960. Dose reconstruction was conducted at other DOE facilities where exposures to offsite residents were lower than the levels recorded for LBNL.

The purpose of positioning a monitoring station at the Olympus Gate location was to provide measurements which would represent a conservative estimate of potential exposures to Bevatron neutron radiation to members of the general public. I expect actual exposures to members of the public to be markedly lower than indicated by the measurements at the Olympus Gate due to: a) increased distance from LBNL accelerators, b) occupancy rates that are much less than 24 hours a day, and c) increased shielding due to the complexity of terrain and building structures.

It is likely that actual exposures received by real persons from the historic release of accelerator neutrons may not be as high as exposures estimated at other DOE facilities where detailed dose reconstruction efforts have been conducted. A recent report (LBNL 45224) by Thomas, Smith and Zeman (2000), which was among the documents reviewed by Franke and Greenhouse and cited in their preliminary review, concluded that the dose from the operation of LBNL accelerators was probably overestimated by a factor of 2 more. This conclusion was based primarily on the substitution of older methods that were standard during the 1950's and 1960's with more modern methods. For real persons who may have been exposed during this time period, these doses would be reduced still further. The locations of actual residences were at distances farther away from the accelerators than the LBNL monitoring stations; there was increased shielding by buildings and terrain, and occupancy times were shorter than 24 hours per day, 365 days per year. It is my understanding that LBNL is currently considering performing more detailed calculations to obtain a realistic estimate of actual offsite exposures. Onsite exposures can be estimated directly from film badges worn by lab employees.

Franke and Greenhouse Comment No. 7.

Based on a review of the tritium sampling plan, the following additions are suggested: (a) the ambient air monitoring network should be expanded of to cover all 16 wind direction sectors (of 22.5° each), (b) the HASL-300 core method should be used for soil sampling, whereby samples would be analyzed for additional depth increments, (c) groundwater sampling would be added in coordination with the State of California Water Resources Board.

The recommendations of Franke and Greenhouse appear to have merit. However, the technical basis for each additional air monitoring station should be clearly established prior to expanding the present network (see my response to Franke and Greenhouse Comment No. 1). Emissions from the NTLF stack are presently monitored in the predominant wind directions. Concentrations in the less frequented wind directions are estimated with mathematical models. These models have shown a tendency to overestimate environmental measurements when the measurements have been averaged over a period of a month to a year. The extent to which these

models overestimate for short-term releases and for wind directions that are not presently monitored remains to be determined.

Expansion of the present system to cover other wind directions may be warranted to respond to Franke and Greenhouse's concern over the potential impact of short term releases at the NTLF. EPA, which determines the adequacy of sampling for Superfund applications, and has indicated that it would be sufficient to expand the present network of ambient monitoring stations by 2 sites. Nevertheless, I believe that public concerns and issues related to short term releases are sufficient to warrant LBNL consideration of additional sampling locations.

I believe that LBNL groundwater sampling is already being coordinated with San Francisco Regional Water Quality Control Board. I am not aware that groundwater contamination from NTLF is a pathway of concern for exposure to LBNL employees or members of the public. Contamination of groundwater may contribute to the presence of tritium in surface waters originating from LBNL site, but the potential dose and health risk from exposure to tritium that has been monitored in surface waters to date are far below limits of concern. Furthermore, surface water sampling is proposed in the Tritium Sampling and Analysis Plan.

The primary pathway of concern regarding releases of tritium from the NTLF is the inhalation and through skin absorption of tritiated water vapor in air. As mentioned previously, the estimated doses based on environmental measurements, limited bioassays, and mathematical modeling of stack emissions have demonstrated that offsite exposures to members of the public are on the order of 100 times less than the 10 mrem/y limit. Further evaluations are being performed to look at possible short-term exposures using the CALPUFF model. These evaluations will consider dispersion under complex terrain and discrete release conditions.

All present soil sampling at LBNL has been performed according to EPA data quality objectives and protocols. Nevertheless, the value gained by using the recommended HASL-300 core sampling method should be evaluated and adopted if the information gained offers a substantial improvement over the current practice at LBNL. Note, however, that EPA only uses information from the top 2 feet of soil to evaluate the potential for contamination of air.

Franke and Greenhouse Comment No. 8.

EPA should provide a parallel calculation for the hazard ranking system if it is assumed that a larger population (such as the full-time equivalent visitor population) is entered for the location of the Lawrence Hall of Science.

This recommendation is valid if CERCLA, and thus the Hazard Ranking System, is applicable to the evaluation of HTO in air resulting from NTLF operations. I do not believe that it is. Releases of tritium from the NTLF are approved under the Clean Air Act (EPA/NESHAPS). I am not aware that CERCLA is applicable for the evaluation of ambient air samples resulting from active operation of the NTLF, which are already approved by EPA.

The Hazard Ranking System of EPA is used to determine eligibility for placing a site on the National Priority List for potential remediation of contaminants in soil, groundwater, and other environmental media. The short residence time of HTO in air means that measured air concentrations of HTO will be directly dependent on ongoing operations at NTLF. Much lower air concentrations would occur from residual amounts of tritium deposited in soil and/or introduced to groundwater. I believe the presence of organically bound tritium in vegetation from past releases from NTLF, and the presence of HTO in groundwater and in soil, would be insufficient to result in an HRS score that would merit consideration for listing on the National Priorities List.

For evaluating the environmental significance of operations of the NTLF, I believe EPA's National Emission Standards for Hazardous Air Pollutants of the Clean Air Act is the relevant legislation to be used, not CERCLA. Compliance with the requirements of the Clean Air Act of EPA depends on estimating the annual dose to a maximally exposed individual offsite. The estimated exposure to the maximally exposed individual is not affected by the estimate of the number of people visiting the Lawrence Hall of Science.

My comments in this correspondence are the result of a preliminary review of the June 30, 2000 report by Franke and Greenhouse. Additional remarks and recommendations may be forthcoming as I have additional time to study the details of their report.

Sincerely,

F. Owen Hoffman, Ph.D.,
President and Director